

REMARKS

As recited in Claim 1, the present invention is drawn to a method of producing a catalyst having a particular formula $P_aMo_bV_cCu_dX_eY_fZ_gO_h$ (wherein X, Y, Z, and a-h are as defined), useful for subjecting methacrolein to vapor phase catalytic oxidization with molecular oxygen, the method comprising mixing

100 parts by mass of a solution or a slurry (liquid A), containing molybdenum atoms, phosphorous atoms and vanadium atoms, in which the content of ammonium species is 0 to 1.5 mol relative to 12 mol of the molybdenum atoms,

5 to 300 parts by mass of a solution or a slurry (liquid B), containing 6 to 17 mol of ammonium species relative to 12 mol of the molybdenum atoms contained in the solution A, and

a solution or a slurry (liquid C), containing an element Z,

wherein the liquid B is **mixed** with the liquid A, the liquid C or a mixture of the liquid A and the liquid C **over a period of 0.1 to 15 minutes**.

(Emphasis added.)

It is understood that the above-emphasized term means that liquid B is **added** over a period of 0.1 to 15 minutes. Once all of liquid B has been added, further stirring may occur, as can be confirmed from Example 1, for example, wherein said addition is carried out in 3 minutes, and stirring the resultant liquid was carried out for 90 minutes. In the discussion below, the terms "mixing" and "adding" are used interchangeably but are understood to be synonymous when referring to time in which mixing or adding is carried out.

The rejection of Claims 1-15 under 35 U.S.C. § 103(a) as unpatentable over JP 2000-296336A (Naito et al), is respectfully traversed.

Naito et al discloses liquids A, B and C, which correspond to those of the present invention. Naito et al's invention is drawn to the mixing order of liquids A, B and C. In Comparative Examples 2 and 3 of Naito et al, the mixing order is changed to $(A+C)+B$ or $A+(B+C)$ and as a result, methacrolein conversion and methacrylic acid yield are reduced compared to Example 1, in which the order is $(A+B)+C$. In Examples 1-4 of the present invention, the mixing order is $(A+C)+B$, similar to Comparative Example 2 of Naito et al. However, the catalysts obtained in these examples are superior in yield of methacrylic acid, in spite of the disclosure of Naito et al.

The Examiner finds that Naito et al does not limit the mixing method. The reason why the addition method is not limited is because Naito et al did not recognize any difference in addition times. However, as demonstrated herein, a superior result is provided, even if a mixing order presumed by Naito et al to be unfavorable is carried out, by adjusting the addition time of B, as claimed herein. In other words, the mixing method influences properties of the catalyst.

Example 11 and Comparative Example 4 in the specification herein provide catalysts having the same composition as the catalyst of Example 8 of Naito et al by using the same raw materials and the same mixing order, i.e., $(A+B)+C$. Comparative Example 4 in the specification herein was conducted by the same procedure as Example 8 of Naito et al and shows the same results as Example 8 in Naito et al. In other words, Comparative Example 4 corresponds to Example 8 of Naito et al. Although Example 8 in Naito et al does not disclose the adding time, it is understood that the adding time is that disclosed for Comparative Example 4 herein. In the comparison between the results of Example 11 and Comparative Example 4, the yield of methacrylic acid increases by 1.7%. In a product like methacrylic acid produced in large quantities, even 1.7% is a very remarkable effect. For the Examiner's benefit, **enclosed herewith** is a partial translation of Example 8, and Table 1 showing

conversion, selectivity and yield data for the Examples and Comparative Examples, of Naito et al.

In addition, the Examiner has not responded to the comparative data in the specification herein, discussed in the previous response. The data is summarized in Table 1 at page 22 of the specification, a copy of which was attached with the previous response. As shown therein, Comparative Examples 1 and 2 are directly comparable to any of Examples 1-3 and 5, the only difference being the liquid B mixing time. As the data show, the selectivity and yield of methacrylic acid was superior using the present invention.

Nor has the Examiner responded to Applicants' previous argument that Naito et al does not recognize that liquid B mixing time is a result-effective variable and therefore, the claims are patentable over Naito et al under the rationale of *In re Antonie*, 559 F.2d 618, 195 USPQ 6, 8-9 (CCPA 1977) (copy of record) (exceptions to rule that optimization of a result-effective variable is obvious, such as where the results of optimizing the variable are unexpectedly good or where the variable was not recognized to be result effective). Applicants are entitled to prevail under either of the above exceptions.

For all the above reasons, it is respectfully requested that this rejection be withdrawn.

The rejection of Claims 1-15 under 35 U.S.C. § 103(a) as unpatentable over U.S. 6,458,740 (Kasuga et al), is respectfully traversed.

The Examiner finds that "Example 1 of Kasuga et al disclose an exemplary embodiment wherein a solution of molybdenum and vanadium components is admixed with phosphoric acid (considered to read upon 'liquid A'), which is then admixed with cesium and nitrate components (considered to read upon 'liquid C'), which is then admixed with an aqueous mixture (considered to read upon 'liquid B')." This finding is clearly erroneous.

The aqueous mixture of which the Examiner finds is "liquid B" is a mixture of liquid A and liquid C (considered to read upon "mixture AC"). There is no disclosure or suggestion

that the mixture AC is admixed with liquid B. Nor does Kasuga et al disclose the period of time in which liquid A is admixed with liquid C. Naturally, there is no disclosure of the mixing time of liquid B.

In addition, the ammonium ion content per 12 mols of molybdenum atoms disclosed in Kasuga et al is a final content. Although the ammonium ion content is more than 0 but not more than 10, the lowest exemplified limit of the content in Kasuga et al is 2.25 mol per 12 mol of molybdenum atoms, as shown in Example 3. On the other hand, the ammonium ion content in liquid A of the present invention is 0-1.5 mol per 12 mol of molybdenum atoms. Particularly, the content in Example 1 herein is 0. That is, in the present invention, molybdenum atom and vanadium atom are admixed with phosphoric acid in a lower ammonium ion content, preferably in the absence of ammonium ion.

Nor, for reasons advanced in the previous response, does Kasuga et al contain any requirement that their catalyst-constituting raw materials be mixed in the form of three liquids, each having particular requirements, let alone that one particular liquid be mixed over a particular period of time. Nor, with regard to the catalyst *per se*, would one skilled in the art, without the present disclosure as a guide, select both (1) copper and (2) potassium, rubidium, or cesium, as the element X in Kasuga et al, and adjust "d" therein to meet the terms of presently-recited "d" and "g" herein.

While the Examiner finds that Applicants' argument that Kasuga et al is much further away from the presently-claimed invention than Naito et al, "does not detract said reference [Kasuga et al] from reading upon the instantly claimed invention," the Examiner has not responded to Applicants' argument that Naito et al is the closest prior art, and over which Applicants have demonstrated patentability, as discussed above. In addition, if Kasuga et al reads on the present invention, why did the Examiner not reject under 35 U.S.C. § 102(b)?

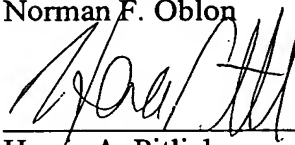
For all the above reasons, it is respectfully requested that this rejection be withdrawn.

Application No. 10/519,407
Reply to Office Action of September 7, 2006

All of the presently-pending claims in this application are now believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

Respectfully submitted,

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Partial translation of JP-A-2000-296336

[0038] [Example 8]

To 400 parts of pure water were added 100 parts of molybdenum trioxide, 8.88 parts of 85% by mass phosphoric acid, 4.74 parts of ammonium metavanadate and 4.11 parts of 60% aqueous arsenic acid solution and stirring was conducted in an autoclave under saturated vapor at 120°C for 3 hours to prepare liquid A. The amount of ammonium species contained in the liquid A was 0.7 mol relative to 12 mols of the molybdenum atoms contained in the liquid A. After cooling the liquid A to 60°C, a solution in which 37.0 parts of ammonium carbonate was dissolved in 80 parts of pure water, which is liquid B was added dropwise to the liquid A and then stirred for 15 minutes to obtain AB mixed solution. The amount of ammonium species in the AB mixed solution was 14 mols relative to 12 mols of the molybdenum atoms. Then, a solution in which 1.40 parts of cupric nitrate was dissolved in 10 parts of pure water, a solution in which 2.34 parts of ferric nitrate was dissolved in 10 parts of pure water and 1.00 part of cerium oxide were added in sequence and with stirring the mixture, liquid C in which 18.0 parts of cesium bicarbonate was dissolved in 30 parts of pure water was added dropwise and stirring was conducted for 15 minutes to prepare a slurry. The slurry thus obtained was heated to 101°C and subjected to evaporation to dryness with stirring. The obtained solid was dried at 130°C for 16 hours. The dried product was subjected to pressing, calcined under nitrogen stream at 400°C for 5 hours and further calcined under air stream at 340°C for 10 hours to obtain a catalyst whose composition was $P_{1.3}Mo_{12}V_{0.7}Cu_{0.1}Fe_{0.1}As_{0.3}Ce_{0.1}Cs_{1.6}$. The results of the reaction conducted using this catalyst in the same manner as in Example 1, except for changing the reaction temperature to 290°C, are shown in Table 1.

[0040]

[Table 1]

	Adding order ¹⁾	Ratio of Ammonium species		Conversion of MAL ³⁾ (%)	Selectivity of MAA ⁴⁾ (%)	Single Current Yield of MAA ⁴⁾ (%)
		Liquid A	AB mixed solution ²⁾			
Ex. 1	(A+B)+C		11.0	83.6	82.4	68.9
Ex. 2	(A+B)+C		6.0	83.0	82.8	68.7
Ex. 3	(A+B)+C		15.0	83.3	82.6	68.8
Ex. 4	(A+B)+C		4.0	80.5	83.1	66.9
Ex. 5	(A+B)+C		20.0	79.8	83.4	66.6
Ex. 6	(A+B)+C		11.0	81.5	82.5	67.2
Comp. Ex.1	(A+B)+C	4.0	7.0	78.6	83.0	65.2
Comp. Ex.2	(A+C)+B		11.0	77.7	83.2	64.6
Comp. Ex.3	A+(B+C)		11.0	78.3	83.0	65.0
Ex. 7	(A+B)+C	0.7	13.3	80.3	83.5	67.1
Ex. 8	(A+B)+C	0.7	13.3	81.2	83.6	67.9
Comp. Ex.4	(A+B)+C	0.7	13.3	76.5	83.4	63.8

1) Means of code in the column of adding order are as follows:

(A+B)+C: Liquid B is added to liquid A and then liquid C is added thereto.

(A+C)+B: Liquid C is added to liquid A and then liquid B is added thereto.

A+(B+C): A mixed solution of liquid B and liquid C is added to liquid A.

(A+B+C): Liquids A, B and C are mixed at one time.

- 2) In the case other than that the adding order is (A+B)+C, ratio of ammonium species of AB mixed solution is that of the mixed solution including liquid C.
- 3) MAL represents methacrolein.
- 4) MAA represents methacrylic acid.